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14. ABSTRACT
The primary aim of this project was to better understand the laser-cluster interaction in the mid-IR range for generating high-order harmonics and evaluate clusters as an alternative source of soft x-rays and attosecond pulses. Clusters as a high-order harmonic generation (HHG) medium was first proposed due to the potential benefits caused by the high local and low global densities by which a large fraction of the incident light can be efficiently stored in the cluster. In fact, early studies claimed that clusters produced higher HHG yield and higher cutoff

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Report Title

Atomic cluster ionization and attosecond generation at long wavelengths

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Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

<u>Received</u> <u>Paper</u>

TOTAL:

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

<u>Received</u> <u>Paper</u>

TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

Number of Presentations: 0.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

<u>Received</u> <u>Paper</u>

TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

<u>Received</u> <u>Paper</u>

TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received		Paper
08/29/2014	1.00	Hyunwook Park, └ Zhou Wang, Hui Xiong, Stephen B. Schoun, Junliang Xu, Pierre Agostini, Louis F. DiMauro. Size Dependent High-order Harmonic Generation in Rare-gas Clusters, P H Y S I C A L R E V I EW L E T T E R S (08 2014)

TOTAL: **1**

Number of Manuscripts:

Books

<u>Received</u> <u>Book</u>

TOTAL:

<u>Received</u> <u>Book Chapter</u>

TOTAL:

Patents Submitted

Patents Awarded

Awards

Graduate Students

NAME	PERCENT_SUPPORTED	Discipline
Zhou Wang	1.00	
FTE Equivalent:	**1.00**	
Total Number:	**1**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
Louis DiMauro	0.10	
Pierre Agostini	0.10	
FTE Equivalent:	**0.20**	
Total Number:	**2**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:...... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):...... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:...... 0.00

Names of Personnel receiving masters degrees

NAME
Total Number:

Names of personnel receiving PHDs

NAME
Total Number:

Names of other research staff

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

Technology Transfer

FINAL PERFORMANCE REPORT

Subject: Final Performance Statement to Dr. Richard Hammond

Contract/Grant Title: Atomic cluster ionization and attosecond generation at long wavelengths.

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Principal Investigators: Louis F. DiMauro (dimauro.6@osu.edu) and Pierre Agostini (agostini.1@osu.edu)

Institution: The Ohio State University, Department of Physics, Columbus, OH 43210

The primary aim of this project was to better understand the laser-cluster interaction in the mid-IR range for generating high-order harmonics and evaluate clusters as an alternative source of soft x-rays and attosecond pulses. Clusters as a high-order harmonic generation (HHG) medium was first proposed due to the potential benefits caused by the high local and low global densities by which a large fraction of the incident light can be efficiently stored in the cluster. In fact, early studies claimed that clusters produced higher HHG yield and higher cutoff energy compared to an atomic target. Accordingly, we have first aimed at verifying the cluster as a "better" source by understanding the HHG mechanism in these nano-solid targets.

For HHG studies, we used a stagnation pressure (*p*)-temperature (*T*) controlled cluster pulsed valve, Even-Lavie ATAD Inc, at the entrance slit of a XUV Hettrick spectrometer. One of the advantages of our source is precise *p*-*T* control for not only producing large (10's of nm) clusters but also to manipulate the atomic density, the number density and the size of the cluster simultaneously. This plays a key role in deconvoluting the various effects contributing to HHG from cluster medium. Another unique feature of our apparatus is a dual gas source in which the cluster valve can be rapidly exchanged with a monomer *cw*-nozzle under vacuum. In this way, HHG signal from the cluster is directly compared to the monomer under the same laser condition thus removing ambiguities in identification.

For the driving fields, we use a Ti:sapphire laser at the wavelength of 800 nm to pump an OPA system, HE-TOPAS from Light Conversion Ltd, covering the mid-IR range. The OPA system produces two wavelength ranges, signal (1.1-1.6 um) and idler (1.6-2.6 um). In addition, the idler is carrier-envelope phase (CEP) stabilized. The total pulse energy is 2.3 mJ (signal + idler), sufficient to produce the desired laser intensity for an argon target.

After characterization of the machine with the target sources, we compared HHG from clusters and monomers. The first question was whether the cluster is a more efficient medium than the monomer for HHG. To address this question, HHG yields in both cluster and monomer targets are studied under the same global atomic density, n_0, using

the dual source. The atomic density was carefully calibrated by measuring the jet parameters and our measurements agreed well with the previously reported values.

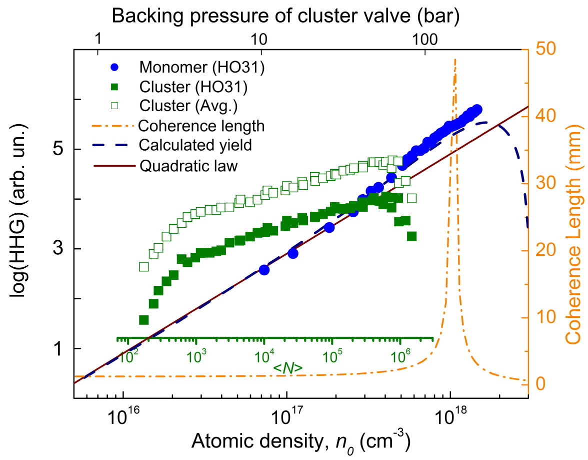

Figure 1: HHG yields in the cluster and monomer and the calculation of the coherent length.

Figure 1 compares the harmonic yields as a function of the atomic density. The general trend from the average yield over all harmonics and specific orders are similar, so here we focus on harmonic-order 31. For proper comparison, the backing pressure (top axis) of the cluster is converted to the atomic density (bottom axis), and also to the average cluster size (green-insert axis) for reference. The monomer's yield are observed to follow a quadratic scaling law (straight line) below 3x10^{17} cm^{-3} while the clusters show a faster initial growth followed by a decreasing rate. Obviously, the HHG yield is larger in the cluster, super-quadratic growth in Fig. 1, at the same atomic density as the monomer, and thus the relative efficiency is indeed higher. Beyond $n_0 = 3\times10^{17}$ cm^{-3} the super-quadratic region in the cluster is different from that of the monomer, which is caused by the fact that the atomic density approaches the maximum coherent length. This behavior in the monomer gas is well reproduced by the calculated phase-matched yield (orange-broken curve) in Fig. 1. The cause of the larger yield will be more discussed below.

We have also measured the HHG group delay (attochirp) for the atom and cluster at a wavelength of 1.3 μm using the RABBITT method. This measurement was only performed using large clusters, ~5 nm in radius, due to the small signal level at lower backing pressures. Figure 2(a) compares the group delay in the monomer and cluster along with the predictions of a semi-classical strong-field model. First, the classical curve agrees well with the measurements except for the dip caused by the Cooper minimum around 50 eV. The positive slope of the attochirp, *e.g.* positive dispersion, is indicative for preferential phase matching of short trajectories for our geometry. Second, the group delay from the cluster and monomer show a striking similarity suggesting that the mechanism for HHG is the same, the 3-step model rather than Bloch model.

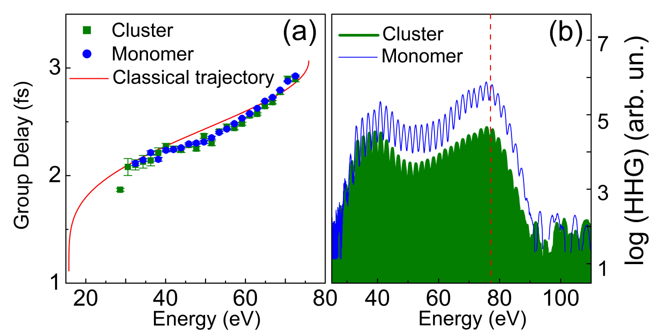

Figure 2: (a) Group delay measurements and (b) HHG spectra comparison.

If the 3-step model were valid for clusters, a question would be whether the returning electron could recombine with a neighbor ion. If so, the cutoff energy of HHG would be extended as proposed by several theoretical models. To study this we compare the harmonic spectra in Fig. 2(b). The two distributions are almost identical showing the same cutoff and agreement with the calculated atomic cutoff (broken line). Again this study suggests that the cluster follows the 3-step model in the same manner as the atom, *i.e.* the returning electron only recombines with the parent ion. Note that the cutoff energy is sensitive to the laser intensity, so our dual source scheme was ideal for this test.

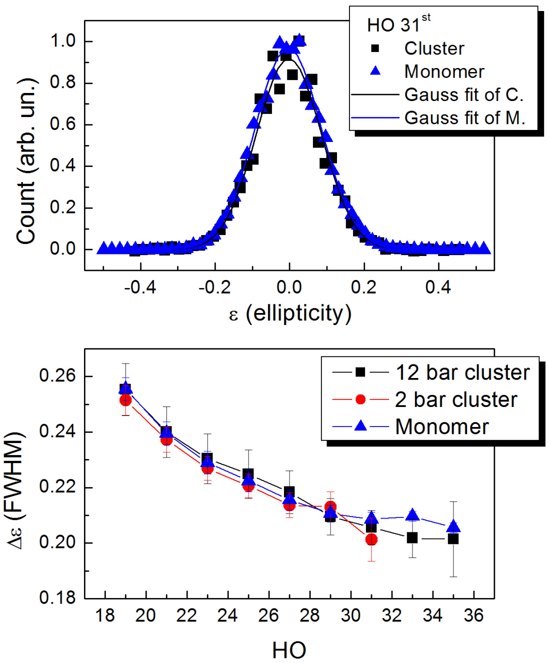

Figure 3: HHG yields in elliptically polarized light: (top) harmonic yield against the ellipticity and (bottom) the sensitivity against the harmonic order.

In order to further investigate this result, we performed an ellipticity study of the HHG yield. The idea is that if the returning electron recombined with a neighbor ion, we should be able to see a target-size effect. For instance, the harmonic yield in the monomer target is very sensitive to the small ellipticity of the driving field due to the modification of the electron trajectory. Figure 3 shows the HHG yield with varying ellipticity for the monomer and, small and large clusters. The top panel shows that the 31^{st}-order harmonic signal decreases with ellipticity the same for the cluster and monomer. The bottom panel summarizes the sensitivity for all harmonic orders. In all cases, the sensitivity to the elliptical light is identical within the error bars regardless of the target size. The similar behavior was also reported in a comparative study between the solid and monomer argon targets.

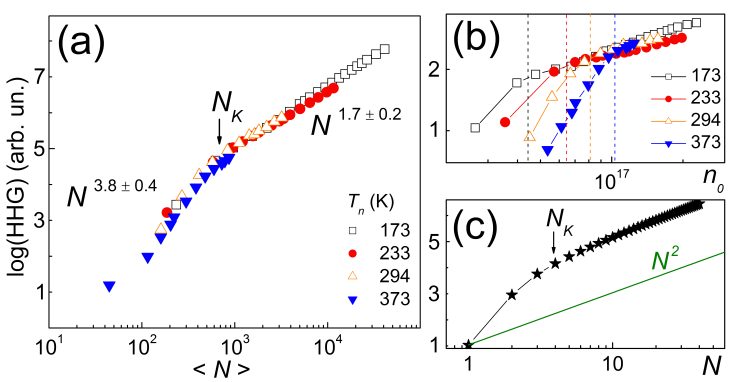

Figure 4: HHG yield for various temperatures and pressures (a) against the cluster size, <N>, and (b) atomic density, n_0. (c) shows the calculated HHG yield as a function of the cluster size.

The results from the group delay, cutoff energy and elliptical dependence agree with the conclusion that the cluster behaves like an ensemble of independent atoms that follow the 3-step model. This is probably a specific characteristic of van der Waals clusters in which the atomic constituents are weakly coupled to each other.

Since many properties of HHG in clusters are similar to

the monomer, then why is the harmonic yield larger in the cluster? From a fundamental perspective, it is interesting to isolate the yield from a single cluster. The main difficulty is to separate the microscopic contribution from the macroscopic one. In this work, precise control of p and T enabled us to extract the single cluster effect. At each T value, p is varied, producing a 2D measurement. From the post analysis of the 2D measurement, we were able to obtain the harmonic yield as a function of cluster size for a single particle, shown in Fig. 4(a). Regardless of combination of p and T, the yield curves show a characteristic knee shape defined by two slopes of $N^{3.8}$ and $N^{1.7}$. The same data is plotted against the atomic density in Fig. 4(b) indicating that this is not a phase matching effect; otherwise the break points would occur at the same atomic density.

We have found that the yield increase is attributed to partial delocalization of electron wave function in the cluster. If delocalization were to simply follow the increase of the size, *i.e.* proportional to N, the yield increase would be constant. A crucial finding of our experiment is the existence of a knee point in the yield, N_K, implying that delocalization increases only up to N_K and remains constant beyond that point.

In order to investigate this we use a modified Lewenstein quantum model in which the cluster is represented by a 1D Coulomb potential for the parent ion and a string of (N-1) Yukawa potentials for the other neutral atoms. The calculations entails a solution of the time-independent Schrödinger equation partially delocalized over only the nearest neighbors which quickly becomes independent of N. Figure 4(c) shows that the calculation qualitatively agrees with the size-dependence observed in the experiment shown in Fig. 4(a). The model also shows a rapid growth in HHG yield, faster than a quadratic (green line) and reproduces the observed knee point.

Finally, we have studied HHG from clusters at various wavelengths. It is found that the harmonic signal falls off rapidly as the wavelength is increased. In fact, the decay rate as a function of wavelength scales as λ^{-12}, which is more rapid than an atomic gas, λ^{-6}.

In summary, within the grant period we have addressed the proposal's aim of understanding laser-cluster interaction for high harmonic generation. The main results have been published in Physical Review Letters.

Archival publications during reporting period: *"Size-Dependent High-order Harmonic Generation in Rare-Gas Clusters"*, Phys. Rev. Lett. **113**, 263401 (2014)

Changes in research objectives: None

Change in AFOSR program manager: None

Extensions granted or milestones slipped, if any: no

New discoveries, inventions or patent disclosures during this reporting period:
The cluster (or plasma) heating mechanism is found to be strongly wavelength dependent, which enables us to better understand the laser-matter interaction (article in preparation).